High harmonic generation in a semi-infinite gas cell

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Abstract: Ten-millijoule 35-femtosecond laser pulses interact with a cell of helium or neon that extends from a focusing lens to an exit foil near the laser focus. High harmonic orders in the range of 50 to 100 are investigated as a function of focal position relative to the exit foil. An aperture placed in front of the focusing lens increases the brightness of observed harmonics by more than an order of magnitude. Counter-propagating light is used to directly probe where the high harmonics are generated within the laser focus. In neon, the harmonics are generated in the last few millimeters before the exit foil, limited by absorption. In helium, the harmonics are produced over a much longer distance.

OCIS codes: (190.4160) Multiharmonic generation.

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1. Introduction

We recently reported on laser high-order harmonic generation in a semi-infinite gas cell, where the gas fills the region from a focusing lens to an exit foil positioned near the focus [1]. The foil partitions the gas-filled region from the vacuum. Figure 1 shows a schematic of the arrangement, which is described in more detail in the next section. In the semi-infinite configuration, we found relatively strong harmonic emission from neon and helium for harmonic orders up to the high eighties. Under our laser conditions, the semi-infinite cell produces markedly brighter harmonics than finite cells with lengths up to 6mm that we tested at various positions near the focus.

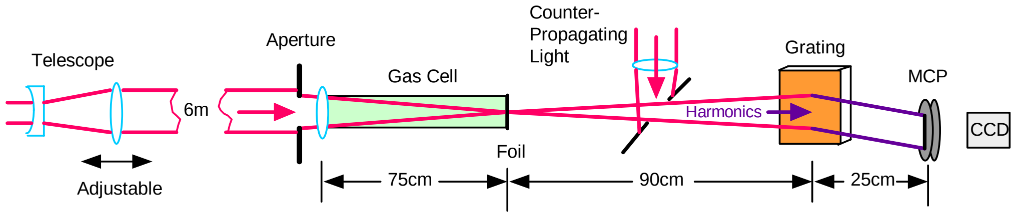

Fig.1. Experimental setup.

The brightness of the high harmonics and the structure of their angular divergences are sensitive to the position of the laser focus relative to the foil, and to the diameter of an aperture placed in the path of the laser before the focusing lens [2].

In this article, we present data scans in movie format showing the dependence of harmonic emission on the diameter of the aperture placed in the laser path. We identify active regions of harmonic production and zones of alternating phase with scans using counter-propagating light as a probe. We also show the dependence of harmonic emission on the laser focus position relative to the exit foil, which we have not previously addressed. The movies offer insight simultaneously into the brightness and the spatial profiles of the harmonic beams as parameters are varied. The results described here are empirical; we defer discussion of specific models describing the phenomena to future works.

Takahashi *et al*. recently reported a conversion efficiency of 1.5×10^{-5} for the generation of the 27^{th} harmonic in an argon-filled cell [3], which is the highest achieved for this harmonic range. They also demonstrated good conversion efficiencies in neon-filled cells, for harmonic orders up to the fifties [4]. In our work, we extend the range of harmonic orders investigated in gas cells up to near one hundred. For these higher orders, significant improvements to phase matching may still be possible. This research compliments work by other groups that have observed strong conversion efficiencies and enhancements in capillaries and gas jets [4-6]. Notably, Gibson and coworkers recently showed enhancement of harmonic orders well beyond a hundred, generated in noble gases and argon ions in corrugated capillary tubes [7-9].

2. Setup

As shown in Fig. 1, the laser pulse enters the gas cell through a 75cm focusing lens. The cell consists of a long glass tube capped with 0.1mm thick molybdenum foil. The harmonics exit through an ~.6mm hole drilled by the laser through the foil. The exit foil of the cell was positioned near the laser focus under vacuum conditions. In the absence of gas, the focal radius was measured to be ~40 μm (within 20% of the diffraction limit) suggesting a peak intensity well above 10^{15} W/cm^2 for our 35 fs ~10 mJ laser pulses. The actual laser spot size and peak intensity when the pulses interact with the gas medium were not determined. Subtle adjustments to a collimating lens located in the beam path well before the chamber permitted the focal position with respect to the exit foil to be varied. These adjustments resulted in slightly different focal spot sizes, which we neglected.

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Counter-propagating light pulses can be introduced into the chamber to probe regions of harmonic emission within the gas cell. A mirror with a small hole drilled through it directed most of the counter-propagating laser energy into the cell while providing an avenue for harmonics to reach the detector. We used a grazing-incidence grating (1200 lines per mm, 2m radius of curvature) to separate and focus individual harmonics onto a micro channel plate coupled to a phosphor screen. A CCD camera simultaneously recorded harmonics orders from the 45th (appearing on the left in each figure) up to near one hundred.

The angular divergence of the harmonics is preserved in the vertical dimension. The full height of all images presented in this article is 6 mrad; most of the harmonics remain within 1-2 mrad divergence angle.

3. Beam aperture effect

Kazamias *et al*. reported marked enhancements to harmonic generation in an argon-filled cell when a partially closed aperture is placed in the path of the laser beam before the focusing lens. They observed enhancements by an order of magnitude for harmonic orders in the twenties. We have also observed the aperture effect in both finite and semi-infinite cells, in our case for higher orders, generated in neon and in helium.

Figure 2 shows high harmonics generated in helium as a function of aperture diameter. Most of the harmonic orders increase strongly when the beam is severely restricted by the aperture. The most pronounced results occur for an aperture diameter near 8mm, compared to the beam diameter of about 12mm. Mid-range harmonics (near 70) increase in brightness by a factor of 15, but higher orders increase less. For example, the 87th harmonic in Helium increases by a factor of 10. Figure 3 shows a scan of harmonics as a function of aperture diameter taken with neon. The behavior is similar to that of helium.

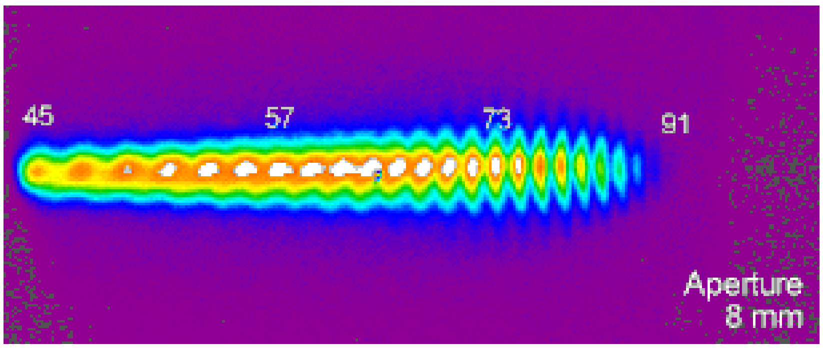

Fig. 2. (1.1 MB) Movie of harmonics generated in 120 torr helium as aperture radius varies.

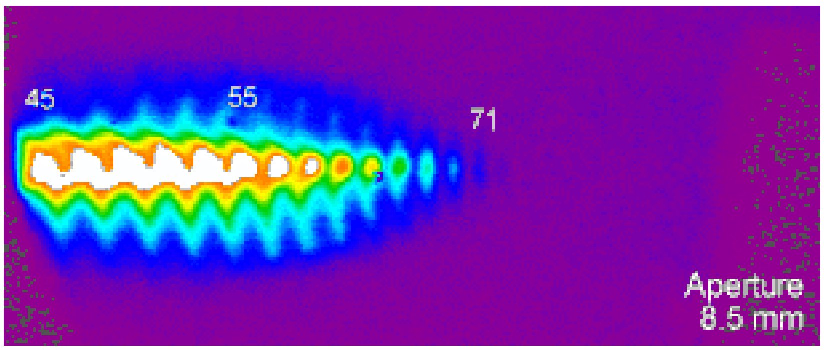

Fig.3. (1.2 MB) Movie of Harmonics generated in 55 Torr neon as aperture radius varies.

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Kazamias et al. identified the effective f-number, the laser-beam spatial quality, and the interplay between the laser phase and the intrinsic phase of high harmonic emission as the primary causes for enhancements to high harmonic signal as the aperture closes. We investigated whether the aperture effect could be explained primarily by a change in the effective f-number of the beam. We installed a longer focal-length lens (and extended the tube on the vacuum chamber), increasing the f-number by 67%. However, this could not reproduce the bright harmonic signal observed with the shorter-focal-length lens and the restricted aperture. Apparently, the aperture introduces phase variations on the laser wave front that are favorable to phase matching.

In the semi-infinite cell, the pulse likely undergoes nonlinear effects such as self-focusing as well as wave-front distortions from ionizing electrons. These have the potential for influencing phase matching, especially over an extended propagation distance as in the semi-infinite cell configuration. Nevertheless, we have observed the aperture effect also in a 1mm-long gas cell, which suggests that conditioning of the beam via propagation within the medium is not critical to the effect. For the scans in Figs. 2 and 3, the laser focus (under vacuum conditions) was positioned near the exit foil of the cell.

4. Effect of focal position relative to foil

When the cell is filled with gas, self-focusing of the pulse pulls the focus inward from the foil by as much as a few centimeters. This is gas-species and pressure dependent. We used the optimal pressure for producing the brightest possible harmonic signal at the best focal setting: 55 torr for neon and 115 torr for helium.

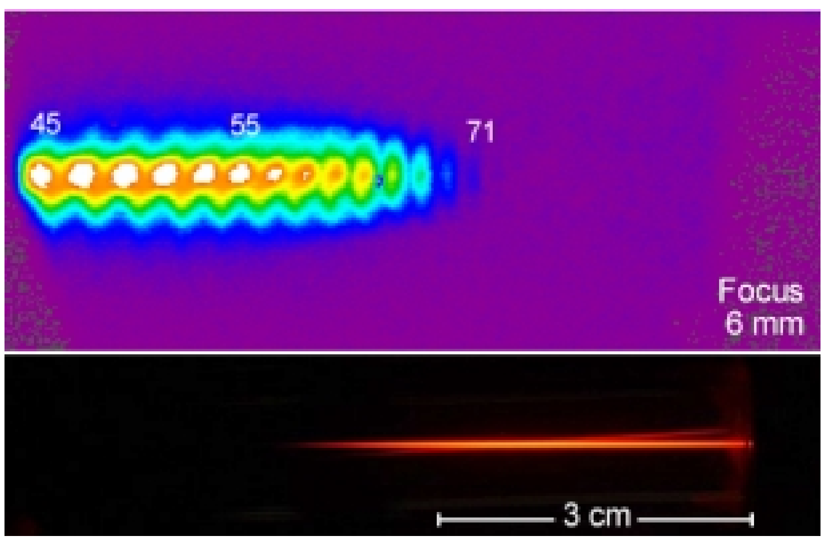

Fig. 4. (.71 MB) Movie of harmonics produced in 55 Torr of neon as focus position is varied with respect to the exit foil. The lower image is a side view of the laser focus.

The harmonic production depends on the position of the laser focus relative to the exit foil. Harmonic production as a function of focal position is shown in Figs. 4 and 5 for neon and helium. The bottom image in each movie is a digital photo of the gas cell taken from the side that shows recombination light from a streak of plasma produced as the laser focuses in the gas. This allows us to see the relationship between the focus and the exit foil. The scale marker is aligned with the foil to mark the position of the end of the cell. The aperture was set at 8 mm for these scans. The similarities between the two gases are evident. Although helium produces significantly higher orders, the evolution of the spatial structure of the emerging beams as a function of focal position looks very much like that for neon. The

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angular divergence, which is fairly narrow, varies with focal position and with harmonic order. For some focal positions, harmonic emission is directed narrowly along the axis within a 1 mrad beam. At other focal positions, emission of some of the higher harmonics forms a 3 mrad ring structure.

Observed harmonic emission comes from a region just inside the exit foil. As demonstrated in the next section, this region is only about 2mm long in neon, but it is considerably longer in helium. Re-absorption of the harmonics limits the amount of emission from regions further within the cell. Thus, as the focal position is scanned, we gain insight into the different phase matching zones along the laser axis.

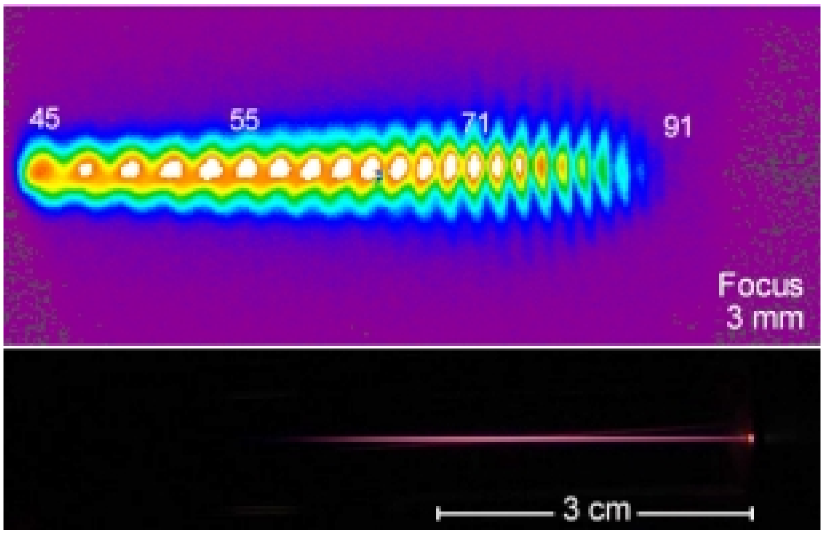

Fig. 5. (.71 MB) Movie of harmonics produced in 115 Torr of helium as focus position is varied with respect to the exit foil. The lower image is a side view of the laser focus.

5. Probing with counter-propagating light

We used counter-propagating light to probe the region near the focus where high harmonics are generated. Even relatively weak counter-propagating light has been demonstrated to suppress harmonic production by interfering with the forward-traveling generating pulse [10]. We used a ~10ps counter-propagating pulse with approximately the same total energy as the forward pulse. The duration of the counter-propagating pulse is sufficient to interact with the forward-going pulse over a distance of 1.5 mm. As the collision zone of the two pulses is swept through the harmonic generation region, variations can be seen in harmonic output.

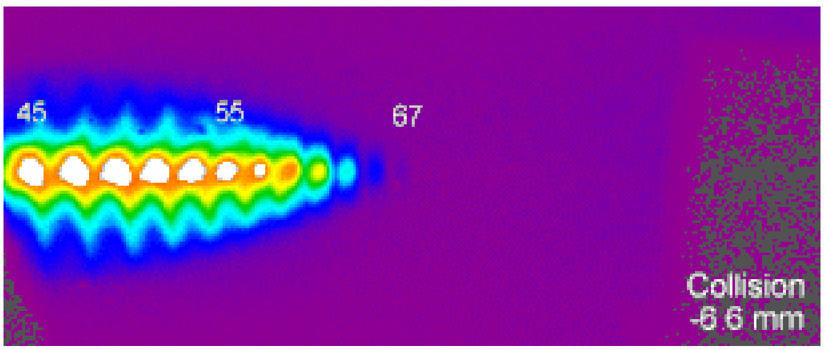

Fig. 6. (1.3 MB) Movie of harmonics in 55 torr neon as counter-propagating light sweeps through the focus.

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Figure 6 shows a scan of neon in a semi-infinite cell at 55 Torr. Initially, the harmonic signal is quite strong, but as the collision zone nears the exit of the cell, suppression is evident. This suggests that most of the harmonics observed in this case are generated in a fairly small (1-2 mm long) region near the foil exit of the cell. Figure 7(a) is a graph of this same scan showing the signal level of several representative harmonics as a function of the position of collision of the pulses. We note that the suppression the harmonics is nearly total and that it takes place over a 2mm distance. We found that shorter counter-propagating pulses could not suppress the emission as completely.

Figure 7 (b) shows a similar scan under conditions where the original signal is weak (neon at 30 torr with the laser focus moved 6mm further in from the foil). Phase matching is poor for these conditions, but it can be improved by suppressing harmonic production in regions with opposing phase [10]. Despite these improvements, we have not yet discovered an enhancement that matches the best harmonics generated under optimal conditions. The graph shows evidence of zones of alternating phase as the harmonic signal peaks and dips.

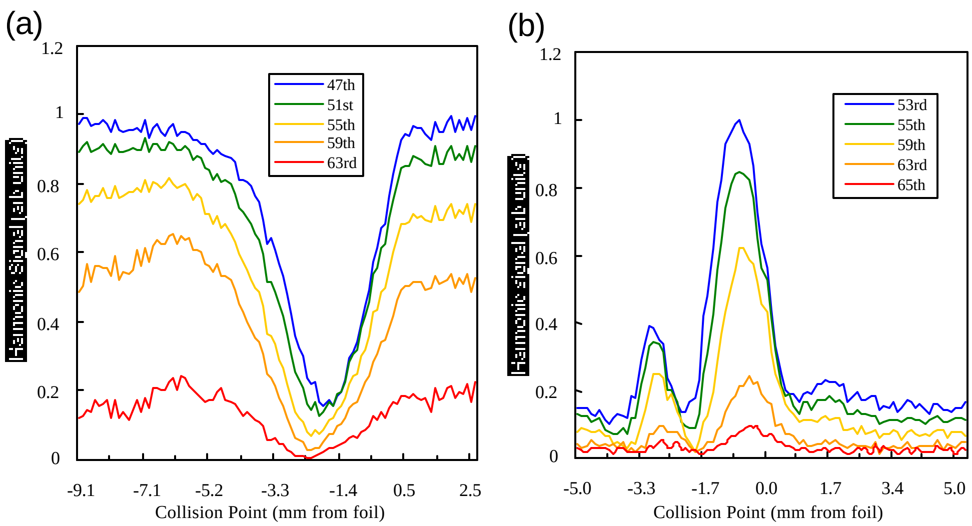

Fig. 7. Graphs of harmonic signal as a fraction of individual maximum vs. counter-propagating pulse collision point. In (b), the focus is 6mm farther upstream and pressure is 25 torr less than in (a), neon at 55 torr.

Probing the harmonic signal generated by helium yields quite different results. We were unable to significantly suppress the harmonic signal with counter-propagating light, even though it interacted with the forward pulse over a 1.5mm distance. This suggests that the harmonics in helium are generated (and phase matched) over a greater distance than in neon.

The difference in absorption by the two gases explains in part the differences in the harmonic generation depths as characterized by the counter-propagating light. Transmission of the 71^{st} harmonic through 2 mm of 55 torr neon is 29%. The 71^{st} harmonic can propagate through 10 mm in helium at 115 torr with the same level of absorption [11].

6. Summary

We have studied harmonic generation in neon and in helium using semi-infinite gas cell geometry. We observed harmonic generation as a function of focal position relative to the cell exit foil. We studied the effect of restricting the laser beam with an aperture at the focal lens. We directly determined where harmonics are produced inside the cell using counter-propagating light.

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Harmonic generation in the two gases shows similar behavior, the most pronounced difference being that helium exhibits stronger overall emission and more orders, up to the low 90s. In the case of helium, the optimal position of focus was 8mm further inside the foil compared to neon (probably due to reduced self focusing). Harmonics from both gases exhibit significant enhancement when the beam is restricted with an aperture at the focusing lens**.** Probing with counter-propagating light revealed that in neon the harmonics are produced in a 2mm region near the exit foil while in helium the harmonics are produced over a much longer distance.

The fact that harmonics are generated in helium over an extended range relative to that in neon is consistent with differences in absorption rates of the two gases. The measurements suggest that the harmonics are phase matched over many millimeters, much longer than would be expected when considering diffraction of the fundamental beam in vacuum. Whether the harmonics are actually phase matched over the entire distance or if emission at different depths is generated at different radii is not determined. In any case, the observations are consistent with those reported by Takahashi et al. who interpreted the behavior as evidence of laser channeling within the gas medium [12].

The scans of harmonics as a function of focal position relative to the exit foil and of aperture radius show variations in angular distribution. These variations may offer clues to the interactions between the laser beam and the generating medium and lead to better understanding of the HHG process. This work is supported by the National Science Foundation under grant number PHY-9985080.

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